Soil and Sediment Monitoring

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Introduction

The soil and sediment surveillance monitoring that Lawrence Livermore National Laboratory performed in 2001 included work in three areas: surface soil in the Livermore Valley and at Site 300, sediment at the Livermore site, and vadose zone soils at the Livermore site.

Soil is weathered material, mainly composed of disintegrated rock and organic material that sustains growing plants. Soil can contain pollutants originally released directly to the ground, to the air, or through liquid effluents. Department of Energy (DOE) guidance for environmental monitoring states that soil should be sampled to determine if there is a measurable, long-term buildup of radionuclides in the terrestrial environment and to estimate environmental radionuclide inventories (U.S. DOE 1991). The guidance recommends monitoring for radionuclides specific to a particular operation or facility as well as those that occur naturally. Particulate radionuclides are of major interest in the LLNL soil monitoring program because airborne particulate releases are the most likely pathway for LLNL-induced soil contamination.

Sediments are defined for the purposes of this chapter as finely divided, solid materials that have settled out of a liquid stream or standing water. The accumulation of radioactive materials in sediment could lead to exposure of humans through their ingestion of aquatic species, sediment resuspension into drinking water supplies, inhalation of dust particles, or as an external radiation source (U.S. DOE 1991). However, the Livermore site and Site 300 do not have habitats for aquatic species that are consumed by people, nor do they have surface drainage that directly feeds drinking water supplies.

Soils in the vadose zone—the region below the land surface where the soil pores are only partially filled with water—are collected in arroyo channels at the Livermore site as part of the Ground Water Protection Management Program (GWPMP). Infiltration of natural runoff through arroyo channels is a significant source of groundwater recharge, accounting for an estimated 42% of resupply for the entire Livermore Valley groundwater basin (Thorpe et al. 1990). Soils in the shallow vadose zone are collected and analyzed to provide information about possible constituents that may be dissolved as runoff water infiltrates through the arroyo to the groundwater.

Sampling Locations

Since 1971, surface soil sampling near the LLNL Livermore site and Site 300 has been part of a continuing LLNL monitoring program designed to measure any changes in environmental levels of radioactivity and evaluate any increase in radioactivity that might have resulted from LLNL operations. These samples have been analyzed for plutonium and gamma-emitting radionuclides, such as depleted uranium used in some explosive tests at Site 300. The inclusion of other gamma-emitting, naturally occurring nuclides (potassium-40 and thorium-232) and the long-lived fission product, cesium-137, provides background information and baseline data on global fallout from historical above-ground nuclear weapons testing. In addition, LLNL analyzes Site 300 soils for beryllium, a potentially toxic metal used at this site. Soils in the Livermore vicinity were analyzed for beryllium from 1991 to 1994. However, analysis for beryllium was discontinued at the Livermore site in 1995, because it was never measured above background values.

Surface soil samples are collected at 19 locations in the Livermore Valley, including 6 sampling locations at the LWRP, an area of known plutonium contamination (**Figure 10-1**) and 14 locations at or near Site 300 (**Figure 10-2**). The locations were selected to represent background concentrations (distant locations unlikely to be affected by LLNL operations) as well as areas where there is the potential to be affected by LLNL operations. Areas with known contaminants, such as the Livermore Water Reclamation Plant (LWRP), are also sampled.

Site 300 soil sampling locations are established around firing tables and other areas of potential soil contamination.

Sediment samples have been collected from selected arroyos and other drainage areas at and around the Livermore site since 1988; these locations (**Figure 10-3**) largely coincide with selected storm water sampling locations (see Chapter 7).

Sediment sampling locations have not been established at Site 300. The drainage courses at Site 300 are steep, causing flowing water to scour the drainages, which prevents the accumulation of sediment. Because of these conditions, sediment sampling at Site 300 is not warranted.

Vadose zone soil sampling has been conducted since 1996. These sampling locations correspond to the same selected storm water sampling locations as the sediment sampling locations (see **Figure 10-3**). Vadose zone samples were not collected in the Drainage Retention Basin because the liner for the basin prevents migration of materials to the groundwater. The collocation of sampling for these three media facilitates comparison of analytical results. As with sediment samples, vadose zone samples are not collected at Site 300.

Approximately 10% of locations are sampled in duplicate; two samples are collected at each location chosen for this sampling. All soil and sediment sampling locations have permanent location markers for reference.

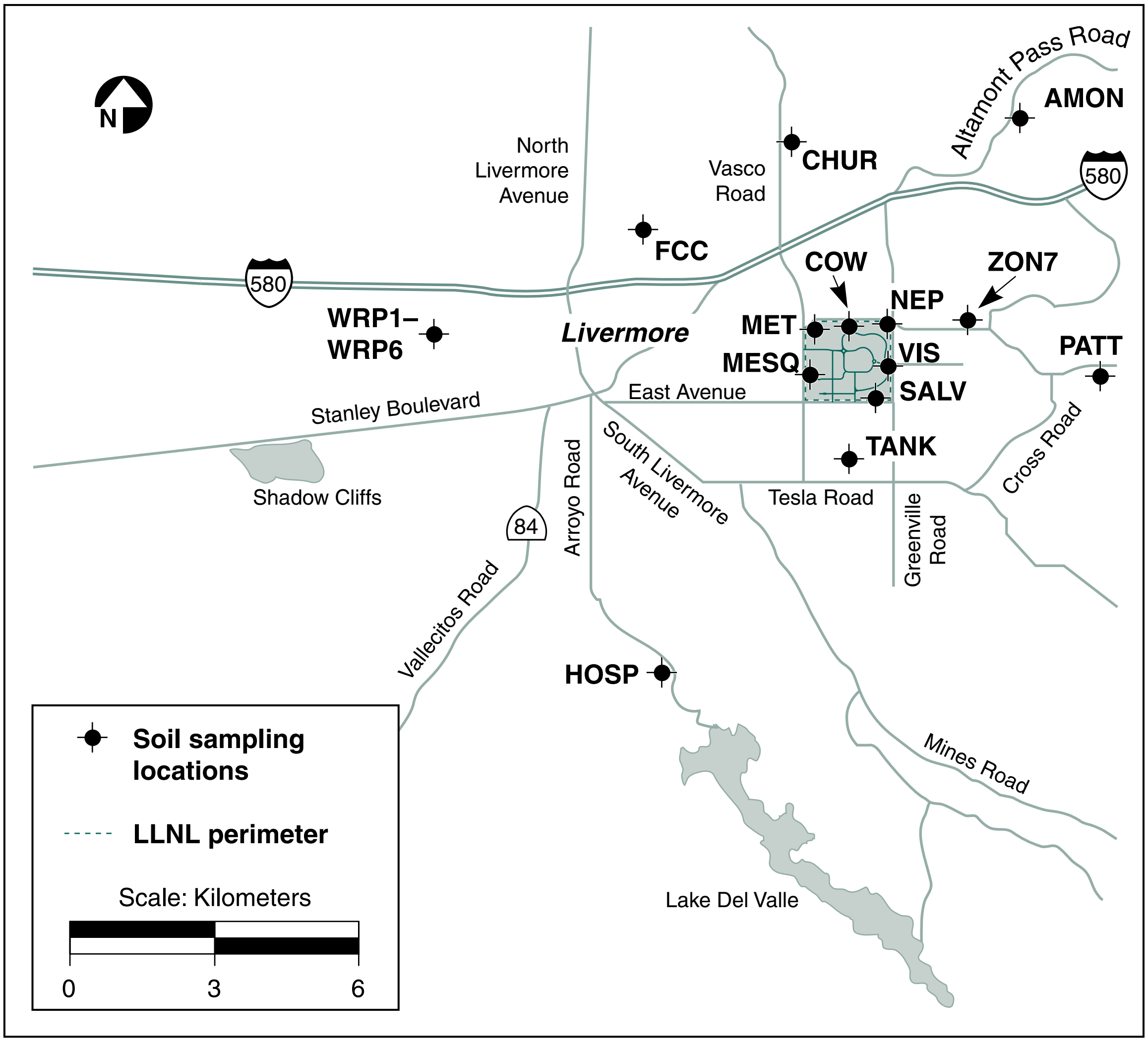

Figure 10-1. Surface soil sampling locations, Livermore Valley, 2001

Methods

Surface soil, sediment, and vadose zone soil sampling is conducted annually according to written, standardized procedures (Tate et al. 1999). Soil samples are collected from undisturbed areas near permanent location markers. These areas are generally level, free of rocks, and unsheltered by trees or buildings. Surface soil samples are collected from the top 5 cm of soil because aerial deposition is the primary pathway for potential contamination, and resuspension of materials from the surface into the air is the primary exposure pathway to nearby human populations.

Sediments are collected annually from drainages at and around the Livermore site after the cessation of spring runoff. Samples to be analyzed for particulate radionuclides are collected from the top 5 cm of soil. Samples to be analyzed for tritium are collected 5–15 cm deep to obtain sufficient water in the sample for analysis. Vadose zone soil samples are collected at 30–45 cm deep for metals analysis

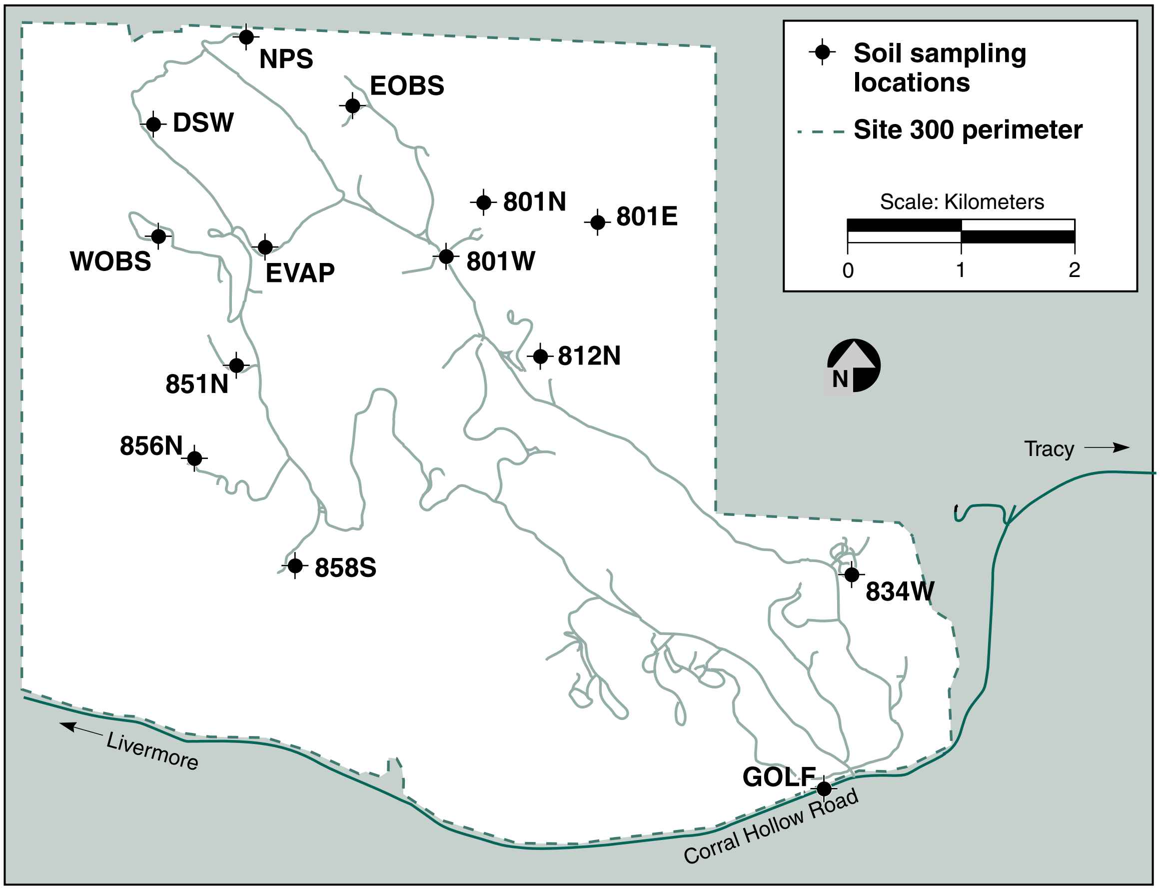

Figure 10-2. Site 300 surface soil sampling locations, 2001

and at 45–65 cm deep for analysis of soluble volatile organic compounds and for polychlorinated biphenyls (PCBs).

In 2001, surface soil samples in the Livermore Valley were analyzed for plutonium and gamma-emitting radionuclides. Samples from Site 300 were analyzed for gamma-emitting radionuclides and beryllium. Analysis of Site 300 soil samples for plutonium was discontinued in 1997 because plutonium has not been used at the site, and sample results have continuously been at background levels since sampling began in 1972. Annual sediment samples collected at the Livermore site were analyzed for plutonium, gamma-emitting radionuclides, and tritium. Vadose zone samples were analyzed for total and soluble metals, and for soluble volatile organic compounds; one vadose zone location was analyzed for PCBs.

Prior to radiochemical analysis, surface soil and sediment samples are dried, ground, sieved, and homogenized. The samples are analyzed by LLNL's Chemistry and Materials Science Environmental Services (CES) laboratory. The plutonium content of a 100-g sample aliquot is determined by alpha spectroscopy. Other sample aliquots (300-g) are analyzed for more than 150 radionuclides by gamma spectroscopy using a high-purity germanium (HPGe) detector (Hall and Edwards 1994a, b, c). The 10-g subsamples for beryllium analyses are sent to a contract analytical laboratory and are

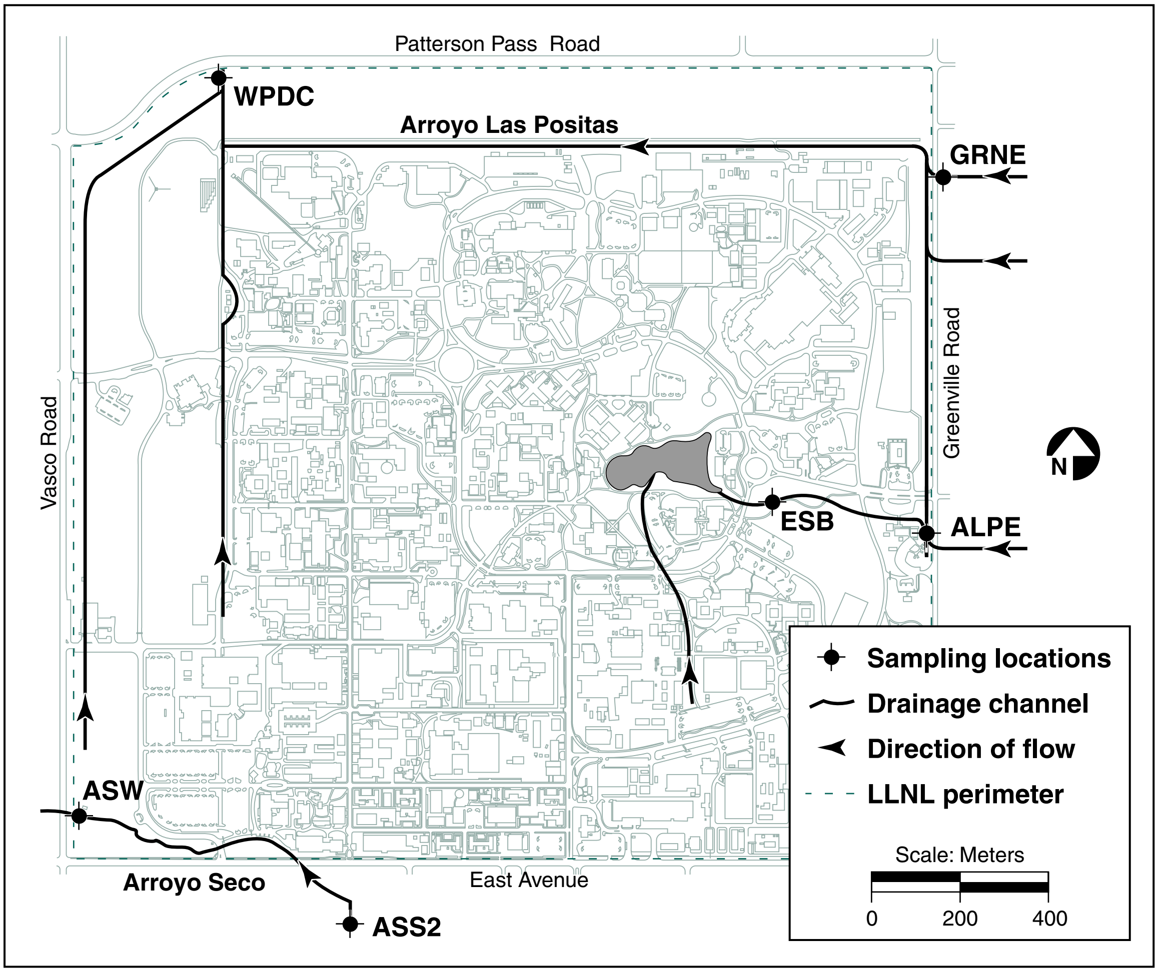

Figure 10-3. Sediment and vadose zone sampling locations on or near the Livermore site, 2001

analyzed by atomic absorption spectroscopy (EPA Method 7091). For sediment samples collected for tritium analyses, CES uses freeze-drying techniques to recover water from the samples and determines the tritium content of the water by liquid-scintillation counting.

Vadose zone soil samples are analyzed by a contract analytical laboratory. The analytical methods include the toxicity characteristic leaching procedure (TCLP, EPA Method 1311) followed by EPA Method 8260 for volatile organic compounds, and total metals by EPA Methods 200.7, 245.2, 7471A, and 6010B. The procedure for determining soluble metals includes the California Waste Extraction Test, followed by the same analytical methods for metals applied to the leachates. In 2001, a vadose zone soil sample from location ESB (Figure 10-3) was also analyzed for PCBs by EPA Method 8082. Chain-of-custody procedures are followed throughout the sampling, delivery, and analytical processes.

Livermore Valley Surface Soil Results

Table 10-1 presents data on the concentrations of plutonium-238 and plutonium-239+240 in the Livermore Valley surface soils. Data for

Table 10-1. Plutonium activity concentrations in Livermore Valley soil, 2001

Location identifier	Plutonium-238 mBq/dry g	Plutonium-239+240 mBq/dry g
L-AMON-SO	0.0019 ± 0.0010	0.073 ± 0.0063
L-CHUR-SO	0.0051 ± 0.0016	0.13 ± 0.0092
L-COW-SO	0.00084 ± 0.00081	0.021 ± 0.0031
L-FCC-SO	0.0016 ± 0.0012	0.066 ± 0.0069
L-HOSP-SO	0.0069 ± 0.0020	0.19 ± 0.012
L-MESQ-SO	0.00084 ± 0.0011	0.033 ± 0.0045
L-MET-SO	0.0017 ± 0.0015	0.047 ± 0.0067
L-NEP-SO	0.0013 ± 0.0010	0.055 ± 0.0054
L-PATT-SO	0.00074 ± 0.0010	0.022 ± 0.0039
L-SALV-SO	0.0063 ± 0.0017	0.066 ± 0.0057
L-TANK-SO	0.0056 ± 0.0019	0.13 ± 0.0097
L-VIS-SO	0.017 ± 0.0032	0.37 ± 0.020
L-ZON7-SO	0.0085 ± 0.0022	0.16 ± 0.011
Median	0.0019	0.066
IQR[a]	0.0050	0.083
Maximum	0.017	0.37

Note: Radioactivities are reported as the measured concentration and either an uncertainty (±2σ counting error) or as being less than or equal to the detection limit. If the concentration is less than or equal to the uncertainty or the detection limit, the result is considered to be a nondetection. See Chapter 14.

a IQR = interquartile range

cesium-137, potassium-40, thorium-232, uranium-235, and uranium-238 in surface soils from the Livermore Valley sampling locations are presented in Table 10-1 of the Data Supplement.

The concentrations and distributions of all observed radionuclides in soil for 2001 are within the ranges reported in previous years and generally reflect worldwide fallout and naturally occurring concentrations.

Plutonium has, in the past, been detected at levels above background at VIS, a perimeter sampling location near the east boundary of the Livermore site. Since 1980, soil samples at this location have generally shown plutonium-239+240 values higher than background. However, in 2001, the measured plutonium-239+240 value for VIS at 370 µBq/dry g (9.93×10^{-3} pCi/dry g) was within the range of background. The slightly higher values at and near the Livermore site have been attributed to historic operations, including the operation of solar evaporators for plutonium-containing liquid waste in the southeast quadrant (Silver et al. 1974). LLNL no longer operates the solar evaporators or engages in any other open-air treatment of plutonium-containing waste. Nonetheless, plutonium-239+240, from historic operations, can be carried off site by resuspension of soil by wind.

Similarly, elevated levels of plutonium-239+240 (resulting from an estimated 1.2×10^9 Bq [32 mCi] plutonium release to the sanitary sewer in 1967 and earlier releases) were first observed in soils near LWRP during the early 1970s, and were again detected at LWRP sampling locations.

As in 1997 through 1999, americium-241 was detected in LWRP samples; it is most likely caused by the natural decay of the trace concentrations of plutonium-241 that were present in the releases to the sewer. Plutonium and americium concentrations for the LWRP are presented in **Table 10-2**. Data for cesium-137, potassium-40, thorium-232, uranium-235, and uranium-238 for LWRP sampling locations are presented in Table 10-1 of the Data Supplement.

Historical plots of median plutonium-239+240 concentrations in soil in the Livermore Valley upwind and downwind of the center of the LLNL Livermore site and at LWRP are shown in **Figure 10-4**. Livermore Valley upwind concentrations have remained relatively constant since monitoring began and generally are indicative of worldwide fallout. Greater variation can be noted in the downwind concentration data, which in 2001 included sampling locations VIS, PATT, NEP, COW, AMON, and ZON7, compared with the upwind data. The concentrations of plutonium at the downwind locations reflect resuspension of low-level plutonium contamination from soils in the southeast quadrant of the Livermore site. Greater variability in plutonium-239+240 is seen in samples from LWRP. Because the plutonium-239+240 is likely to be present in discrete particles, the random presence or absence of the particles dominates the measured plutonium-239+240 in any given sample.

Table 10-2. Plutonium and americium activity concentrations in LWRP soil, 2001

Location identifier	Plutonium-238 mBq/dry g	Plutonium-239+240 mBq/dry g	Americium-241 mBq/dry g
L-WRP1-SO	0.34 ± 0.017	6.5 ± 0.23	4.0 ± 2.8
L-WRP2-SO	0.20 ± 0.013	3.4 ± 0.13	1.5 ± 0.83
L-WRP3-SO	0.054 ± 0.055	1.0 ± 0.043	<0.68
L-WRP4-SO	0.017 ± 0.0030	0.32 ± 0.017	<0.54
L-WRP5-SO	0.085 ± 0.0078	1.9 ± 0.078	<0.53
L-WRP6-SO	0.061 ± 0.0063	1.1 ± 0.047	<0.51
Median	0.073	1.5	<0.61
IQR[a]	0.12	2.0	Not calculated[b]
Maximum	0.34	6.5	4

Note: Radioactivities are reported as the measured concentration and either an uncertainty (±2σ counting error) or as being less than or equal to the detection limit. If the concentration is less than or equal to the uncertainty or the detection limit, the result is considered to be a nondetection. See Chapter 14.

a IQR = interquartile range

b Interquartile range not calculated because of high incidence of nondetections.

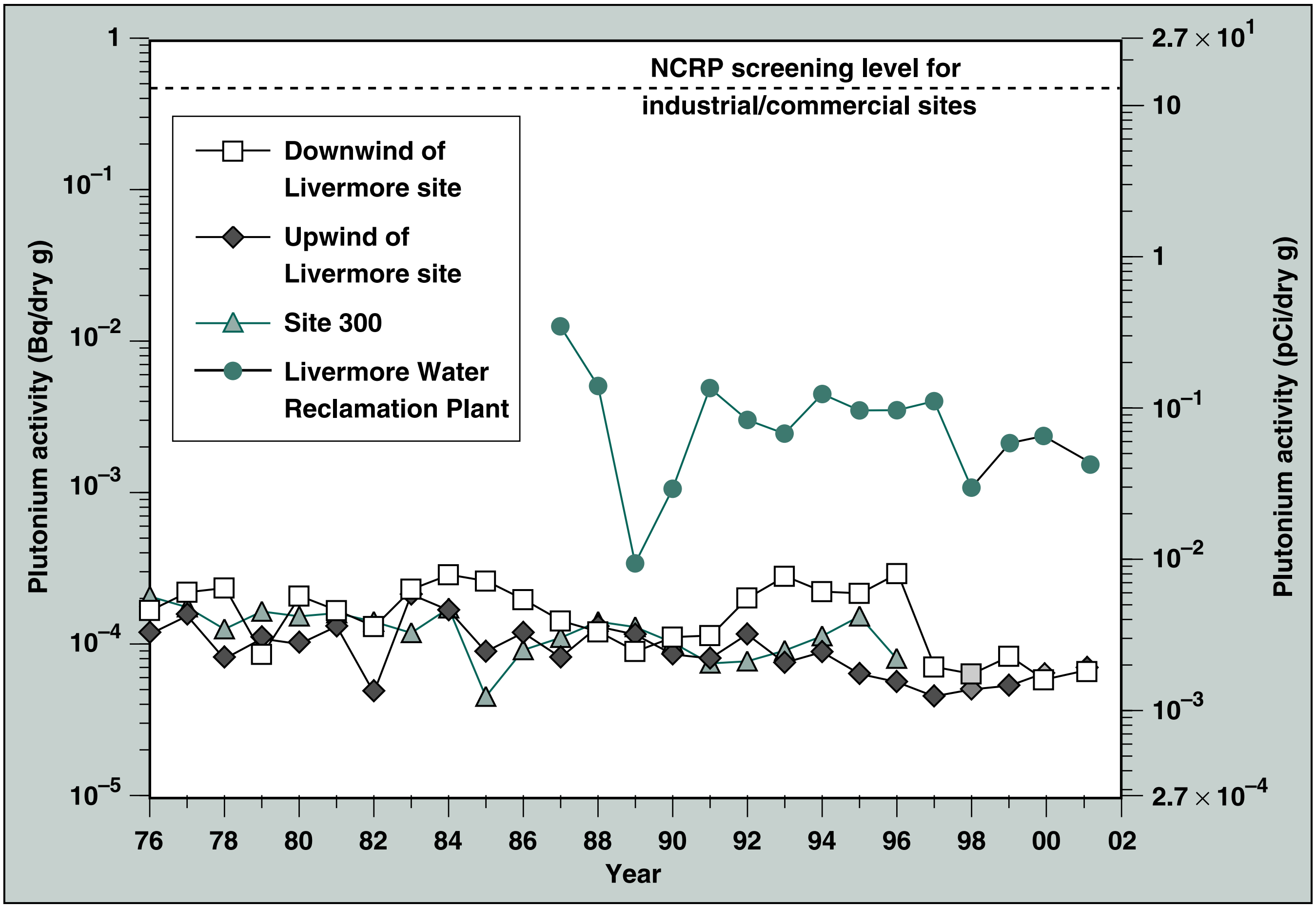

Figure 10-4. Median plutonium-239+240 activities in surface soils, 1976–2001. Upwind and downwind designations are relative to the center of the Livermore site.

Livermore Site Sediment Results

Table 10-3 presents data for plutonium-238, plutonium-239+240, and tritium in sediment samples. Data for cesium-137, potassium-40, thorium-232, uranium-235, and uranium-238 for surface sediment sampling locations are presented in Table 10-1 of the Data Supplement. The levels of plutonium-239+240 were generally at background concentrations, reflective of worldwide fallout. Sampling location ESB (see **Figure 10-3**) shows a moderately higher value for plutonium than values at other locations. The value may be attributed to historic actions because this location is in a drainage area for the southeast quadrant at LLNL. Tritium concentrations were within the range of previous data. The highest detected value, 19 Bq/L (520 pCi/L), was also at location ESB. Location ESB is not only located in a settling basin that serves to remove particles from the surface water drainage of the southeast quadrant before the water enters the Drainage Retention Basin (DRB), but is also located very near the DRB. The DRB contains water with similar concentrations of tritium (see Chapter 7). The detection at ESB is 3% of the drinking water standard of 740 Bq/L (20,000 pCi/L) for tritium. Tritium in sediments will continue to be evaluated as long as the measured values remain above the detection limits of the liquid scintillation analytical method. As for

Table 10-3. Plutonium and tritium activity concentrations in surface sediment, 2001

Location identifier	Plutonium-238 mBq/dry g	Plutonium-239+240 mBq/dry g	Tritium Bq/L
L-ALPE-SD	0.0015 ± 0.00097	0.021 ± 0.0033	1.4 ± 2.0
L-ASS2-SD	0.00094 ± 0.0013	0.0094 ± 0.0031	0.71 ± 1.9
L-ASW-SD	0.00092 ± 0.0011	0.013 ± 0.0028	0.2 ± 2.0
L-ESB-SD	0.22 ± 0.014	1.9 ± 0.079	19 ± 2.7
L-GRNE-SD	0.003 ± 0.0014	0.035 ± 0.0046	1.2 ± 2.0
L-WPDC-SD	–0.000058 ± 0.00058	0.0091 ± 0.0023	2.5 ± 2.0
Median	0.0012	0.017	1.3
IQR[a]	0.0017	0.021	1.4
Maximum	0.22	1.9	19

Note: Radioactivities are reported as the measured concentration and either an uncertainty (±2σ counting error) or as being less than or equal to the detection limit. If the concentration is less than or equal to the uncertainty or the detection limit, the result is considered to be a nondetection. See Chapter 14.

a IQR = interquartile range

surface soil, the concentrations and distributions of all observed radionuclides in surface sediment for 2001 are within the ranges reported in previous years and generally reflect worldwide fallout and naturally occurring concentrations.

Livermore Site Vadose Zone Soil Results

Analytical results for vadose zone soil samples are compared with soil reuse standards developed by LLNL and approved by the San Francisco Bay Regional Water Quality Control Board (SFBRWQCB) (Folks 1997; Marshack 2000). Metals background concentrations are based on naturally occurring levels in the soil, considering first the results for total metals and then the soluble metals test. Natural background levels for organic compounds and tritium at this depth are zero, or below detectable levels. Soils containing materials at levels above background still may not adversely affect the groundwater. If there are any detected organic compounds or tritium, the designated level methodology (DLM) (i.e., application of a simple attenuation factor and specific water quality objectives) is used to determine the soluble levels of contaminants that would not adversely impact groundwater beyond its beneficial uses. (Background and DLM de minimis values are presented in Tables 10-3 and 10-4 in the Data Supplement.)

All analytical results for soluble VOCs were below detection limits. Unfortunately, detection limits were elevated for all compounds due to matrix interferences. All total metals concentrations were within site background. See Tables 10-5 to 10-7 in the Data Supplement for analytical results for VOCs and metals. A PCB, Arochlor 1260, was detected at 2.1 mg/kg at location ESB. The presence of PCBs suggests that this sample represents residual low-level contamination from the 1984 excavation of the former East Traffic Circle landfill (see Chapter 9). The detected concentrations are below the federal and state hazardous waste limits. Tritium results from the sediment sampling were evaluated by the DLM method and were all below de minimis levels (see **Table 10-3**).

Site 300 Results

Table 10-4 presents data on the concentrations of uranium-235, uranium-238, and beryllium in soil from the Site 300 sampling locations; 2001 soils data for Site 300 for cesium-137, potassium-40, and thorium-232 are found in Table 10-2 of the Data Supplement. The concentrations and the distributions of all observed radionuclides in Site 300 soil for 2001 lie within the ranges reported in all years since monitoring began. The ratio of uranium-235 to uranium-238 generally reflects the natural ratio of 0.7%. There is significant uncertainty in calculating the ratio, however, due to the difficulty of measuring low activities of uranium-238 by gamma spectrometry. Historical trends of uranium-238 concentrations from both the Livermore Valley and Site 300 are shown in Figure 10-5. Median values have remained relatively constant for both places. The highest values at Site 300 result from the use of depleted uranium in explosive experiments.

Table 10-4. Uranium and beryllium concentration in Site 300 soil, 2001

Location identifier	Uranium-235[(a)] μg/dry g	Uranium-238[(b)] μg/dry g	Uranium-235 and Uranium 238 ratio	Beryllium mg/kg
3-801E-SO	0.020 ± 0.0092	1.5 ± 1.1	0.013 ± 0.011	0.8
3-801N-SO	0.036 ± 0.012	9.2 ± 0.22	0.0039 ± 0.0013	1.4
3-801W-SO	0.020 ± 0.0090	4.0 ± 0.79	0.0050 ± 0.0025	0.9
3-812N-SO	0.042 ± 0.0084	18 ± 2.0	0.0023 ± 0.00053	1.0
3-834W-SO	0.022 ± 0.012	1.7 ± 1.3	0.013 ± 0.012	1.4
3-851N-SO	0.029 ± 0.0092	2.6 ± 0.78	0.011 ± 0.0048	1.1
3-856N-SO	0.016 ± 0.0081	1.8 ± 1.1	0.0089 ± 0.0071	1.1
3-858S-SO	0.024 ± 0.012	2.0 ± 1.2	0.012 ± 0.0094	0.8
3-DSW-SO	0.019 ± 0.0074	2.3 ± 0.74	0.0083 ± 0.0042	0.8
3-EOBS-SO	0.019 ± 0.011	1.7 ± 0.99	0.011 ± 0.0090	1.1
3-EVAP-SO	0.024 ± 0.0095	4.0 ± 0.79	0.0060 ± 0.0027	0.7
3-GOLF-SO	0.020 ± 0.0089	1.6 ± 0.78	0.013 ± 0.0086	0.8
3-NPS-SO	0.023 ± 0.0092	2.0 ± 0.79	0.012 ± 0.0067	0.7
3-WOBS-SO	0.017 ± 0.01	1.8 ± 0.99	0.0094 ± 0.0076	0.8
Median	0.021	2.0	0.010	0.85
IQR[(c)]	0.0048	1.9	0.0054	0.3
Maximum	0.042	18	0.013	1.4

Note: Radioactivities are reported as the measured concentration and either an uncertainty (±2σ counting error) or as being less than or equal to the detection limit. If the concentration is less than or equal to the uncertainty or the detection limit, the result is considered to be a nondetection. See Chapter 14.

a Uranium-235 activities can be determined by multiplying the mass concentration provided in the table in μg/dry g by specific activity of uranium-235, i.e., 0.080 Bq/μg, or 2.16 pCi/μg.

b Uranium-238 activities can be determined by multiplying the mass concentration provided in the table in μg/dry g by specific activity of uranium-238, i.e., 0.01245 Bq/μg, or 0.3367 pCi/μg.

c IQR = interquartile range

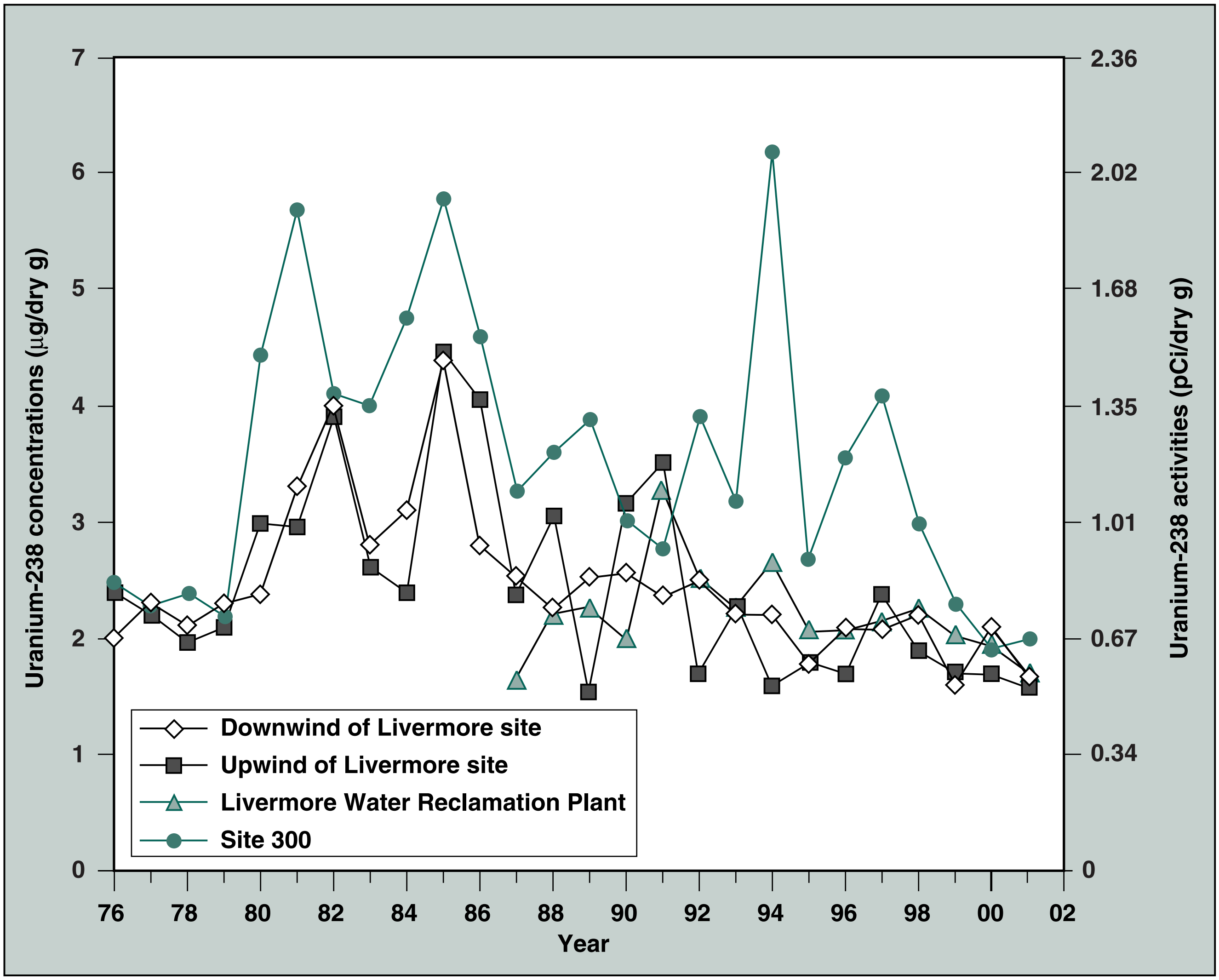

Figure 10-5. Median uranium-238 concentrations in surface soils, 1976–2001. Upwind and downwind designations are relative to the center of the Livermore site.

Environmental Impact

This section discusses the environmental impact of operations at the LLNL Livermore site and Site 300 inferred from soil, sediment, and vadose zone soil monitoring.

Livermore Site

Routine surface soil, sediment, and vadose zone soil sample analyses indicate that the impact of LLNL operations on these media in 2001 has not changed from previous years and remains insignificant. Most analytes of interest or concern were detected at background concentrations or in trace amounts, or could not be measured above detection limits.

The highest value of 6.5 mBq/dry g (0.18 pCi/dry g) for plutonium-239+240 measured at LWRP is 1.4% of the National Council on Radiation Protection (NCRP) recommended screening limit of 470 mBq/g (12.7 pCi/g) for property used for commercial purposes (NCRP 1999). Statistical analysis shows no general increase or decrease in plutonium-239+240 values with time.

Over the years, LLNL has frequently investigated the presence of radionuclides in local soils. Several of the studies are listed in **Table 10-5**. LLNL sampling of surface soil, sediment, and vadose zone soil will continue on an annual basis.

Site 300

The concentrations of radionuclides and beryllium observed in soil samples collected at Site 300 are within the range of previous data and are generally representative of background or naturally occurring levels. The uranium-235/uranium-238 ratios that are indicative of depleted uranium occur near active and inactive firing tables at Buildings 801 and 812. They represent a small fraction of the firing table operations that disperse depleted uranium. The uranium-238 concentrations are below the NCRP recommended screening level for commercial sites of 313 µg/g (3.9 Bq/g or 105 pCi/g). Historically, some measured concentrations of uranium-238 near Building 812 have been greater than the screening level. The investigation and characterization planned for the area surrounding Building 812 will clarify the nature and extent of the contamination in the area. An investigation of the groundwater near Building 812 is in progress. The groundwater has been found to contain depleted uranium. For a further discussion of this investigation, see Chapter 8.

Table 10-5. Special soil studies

Year	Subject	Reference
1971-1972	Radionuclides in Livermore Valley soil	Gudiksen et al. 1972; Gudiksen et al. 1973
1973	Radionuclides in San Joaquin Valley soil	Silver et al. 1974
1974	Soil study of southeast quadrant of Livermore site	Silver et al. 1975
1977	Sediments from LLNL to the San Francisco Bay	Silver et al. 1978
1980	Plutonium in soils downwind of the Livermore site	Toy et al. 1981
1990	195 samples taken in southeast quadrant for study	Gallegos et al. 1992
1991	Drainage channels and storm drains studied	Gallegos 1991
1993	EPA studies southeast quadrant	Gallegos et al. 1994
1993	Historic data reviewed	Gallegos 1993
1995	LLNL, EPA, and DHS sample soils at Big Trees Park	MacQueen 1995
1999	Summary of results of 1998 sampling at Big Trees Park	Gallegos et al. 1999
2000	Health Consultation, Lawrence Livermore National Laboratory, Big Trees Park 1998 Sampling	Agency for Toxic Substances Disease Registry 2000
2002	Livermore Big Trees Park:1998 Results	MacQueen et al. 2002